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PRELIMINARY STUDIES OF NF_3 +H_2 PRODUCTION OF $N_2(A^3 \Sigma^+ U)$ IN A SUPERSONIC FLOW

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February 1988

Final Report

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AIR FORCE WEAPONS LABORATORY
Air Force Systems Command
Kirtland Air Force Base, NM 87117-6008

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This final report was prepared by the Air Force Weapons Laboratory, Kirtland Air Force Base, New Mexico, Job Order 33260385. Capt Nanette D. Founds (ARBL) was the Laboratory Project Officer-in-Charge.

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09	03		

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

a 1 Delta, b 1 Sigma

The N_2F_4 has been used in reaction with H_2 and D_2 to produce $NF(a^1\Delta)$, $NF(b^1\Sigma)$ and $N_2(A^3\Sigma^+u)$. The N_2F_4 is shock sensitive and not commercially produced. Nitrogen trifluoride, NF_3 has been used in HF lasers as a fuel. By operating a conventional HF laser combustor at lower temperatures and less fuel, this study was designed to investigate the use of NF_3 as a replacement for N_2F_4 in $NF(a^1\Delta)$ and $N_2(A^3\Sigma^+u)$ production. Both of these energy transfer agents are of interest in the area of chemical lasers as an energy pump for a suitable species such as IF or NO. The NF_3 was not only used in the combustor and injected into an H_2 stream, but was also injected directly into the flow field. Keywords: Metastable species, Excitation, Activation, Nitrogen Fluorides.

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1.0 INTRODUCTION

The reaction between N_2F_4 and H_2 has been studied extensively (Refs. 1-4). Because of the hazard of working with the shock-sensitive N_2F_4 and the lack of commercial production of N_2F_4 an alternate means of producing the required NF_2 was investigated. The NF_3, nitrogen trifluoride, has been used frequently as a fluorine source for hydrogen fluoride (HF) and deuterium fluoride (DF) lasers.* The handling of ND_3 is substantially easier than N_2F_4 (Refs. 5,6). The NF_3 combustor method used in HF(DF) lasers to produce F atoms was assumed to work, except that a lower temperature in the combustor would be required to retain the NF_2 intact. Combustion of the NF_3 should provide the NF_2 and F source for the following reactions to produce $NF(a^1\Delta)$ and $N_2(A^3\Sigma)$.

$$NF_3 \xrightarrow{\Delta} NF_2 + F \tag{1}$$

$$H_2 + F \rightarrow H + HF \tag{2}$$

$$NF_2 + H \rightarrow NF(a^1\Delta) + HF \tag{3}$$

$$NF(a^1\Delta) + H \rightarrow N(^2D) + HF \tag{4}$$

$$NF(a^1\Delta) + N(^2D) \rightarrow N_2(B) + F \tag{5}$$

$$N_2(B) \rightarrow N_2(A) + h\nu \tag{6}$$

The $NF(a^1\Delta)$ and $N_2(A)$ have been considered as energy storage molecules to be used in the transfer of energy to atoms or molecules suitable for lasing. Both $NF(a^1\Delta)$ and $N_2(A)$ have long lifetimes which makes them unsuitable for lasing (Refs. 7,8).

*Communications with operators of the RACHL device at AFWL and Mr. Chuck Lorenzen, Rocketdyne, KAFB, NM.

2.0 DEVICE

The experimental device has been described in Ref. 9; however, Fig. 1 is provided to show the overall layout. The NF_3 was injected where the fluorine port is indicated for the first series of tests. The later test series involved injecting NF_3 through the trip jets directly into the cavity. The device was constructed of 316L stainless steel. All flow systems were made of stainless steel because of the corrosive nature of the gases.

The nozzle used was the BCL-16. The supersonic nozzle has been studied for HF/DF laser applications (Ref. 10). The nozzle assembly consisted of a combustor section leading into the primary jets. The combustor portion of the nozzle assembly was operated (as it had been designed) to produce F atoms. The hydrogen or deuterium and flourine or NF_3 were injected into the combustor along with helium diluent at a molar ratio of F_2: D_2: He: NF_3 of approximately 3: 5.5: 1: 4.3 to begin testing, and was then optimized.

A one-half cross section of the nozzle is shown in Fig. 2. The nozzle is symmetric in the X-Y plane about the indicated X-axis. The He purge flow as indicated in Fig. 2 represents the He bleed plate which was an annular injector positioned on the gas input wall of the device. The bleed plate injection was used to confine the nozzle flame and kept the observation windows from direct contract with the flame. The BCL-16 contains three secondary nozzles through which either H_2 or D_2 could be mixed with the F atoms arriving through the two primary nozzles. Using NF_3 in the combustor involved starting the combustion with F_2 + D_2 and then mixing in NF_3. At lower NF_3 flow rates, a constant low flow of F_2 was required for sustained combustion.

3.0 DIAGNOSTICS

3.1 NF($a^1\Delta$) AND NF($b^1\Sigma$) DIAGNOSTICS

The NF($a^1\Delta$) diagnostic was an important part of the reaction analysis. The overall arrangement of the NF(a) and NF(b) diagnostics is shown in Fig. 3 and has been described in Ref. 11. The diagnostic as applied to the device is shown in Fig. 4. Figure 5a. shows the result of a digitized photograph of the NF_3 flame. The flame shape was less broadened than the N_2F_4 flame because of lower temperatures in the flow. Figure 5b illustrates the comparison. The actual width of the flame was used to determine the volume viewed by the diagnostic along the path of the scan. The spatial filter was mounted on a remotely operated translation stage with a linear voltage displacement transducer to accomplish scans across the flow field of the device with a known position. Sample scans of the NF($a^1\Delta$) and NF($b^1\Sigma$) emissions are shown in Figs. 6 and 7.

Errors for the diagnostics were based upon the extent of interferences from other emissions and calibration errors. The error for the NF($b^1\Sigma$) diagnostic was determined to be ±10% with a range to 10^{11} to 10^{13} molecules/cm^3. For the NF($a^1\Delta$) diagnostic, the error was larger due to the interferences from other emissions in the system and was estimated at ±20% with a range of 10^{14} to 10^{16} molecules/cm^3.

3.2 OPTICAL MULTICHANNEL ANALYZER (OMA)

The OMA III 1460R system (EG&G PAR) was used to monitor the change in emission over a wide wavelength range (usually 300-900 nm) at a fixed point within the device. The OMA III system consisted of a nonintensified diode array head (Model 1412) coupled to a Model 1233 polychromator. The triple grating polychromator was operated using the 150 l/mm or 600 l/mm grating. The emission from the device was delivered to the poylchromator via a fused silica fiber optic matched to the entrance slit. The system using the 150 l/mm grating had a wavelength resolution of 0.6 nm/channel. Using the fiber optic with a spatial filter, the spatial resolution was about 4 cm.

4.0 OPTIMIZATION OF $N_2(B)$

4.1 COMBUSTOR INJECTION OF NF_3

The combustor flow rates were varied to obtain an intense $N_2(B)$ visible spectra as a method of tracking $N_2(A)$ production. After the combustor was optimized, the secondary H_2 was varied. Figures 8 through 11 show some of the results of the parametric NF_3 studies. The $N_2(B)$ population was not as sensitive to NF_3 flow as might be expected. Table 1 summarizes several test conditions where high $N_2(B)$ levels were achieved. The $N_2(B)$ levels were as high as similar tests using N_2F_4. The $NF(a^1\Delta)$ production was lower than on tests with N_2F_4. A sample OMA III scan is shown in Fig. 12, using NF_3. A scan using N_2F_4 is shown for comparison in Fig. 13. The feature which is most striking is a peak around 670 nm. The possible problem is that the second order of the NH peak is causing the visible peak; however, the scans were also performed using ultraviolet blocking filters and the peak remained. The identity of the peak has not been determined. The marked lack of $NF(a^1\Delta)$ and $NF(b^1\Sigma)$ when NF_3 is used is interesting in that large $N_2(B)$ populations were still found. One conclusion may be that the reaction is occurring more rapidly in the NF_3 combustion. This is probably due to more complete mixing occurring between the NF_2 primary and H_2 secondary jets; although to confirm this, a mixing study should be performed. The lack of definition of the features in $N_2(B\text{-}A)$ series in Fig. 12, may be due to greater broadening caused by slightly higher pressure in the NF_3 experiment.

4.2 TRIP JET INJECTION OF NF_3

Based upon the high temperature in the cavity with N_2F_4 and the known reaction of NF_3 with H_2, injection of NF_3 directly into the cavity was tried. Visible emission was seen via video cameras focused on the device. The measured emission using the NF(a) and NF(b) diagnostics were much lower than the NF_3 combustor studies. The NF(a) was two orders of magnitude lower and the NF(b) not detectable on several tests. Sample OMA III scans are shown in Figs. 14

to 17. Very little N_2(B-A) is shown and mainly the HF ($\Delta v = 4$) sequence bands are apparent. The scans are at increasing distance from the NEP or the point of injection. By Fig. 17, one observes an increase in the N_2(B-A) emission. This corresponds to 9 cm from the NEP. The N_2(B-A) emission may increase downstream; however, the reaction of $NF_3 + H_2$ and the mixing combined is too slow for the supersonic application. More investigation of the system may provide a method for accelerating the reaction. Mixing can be improved by a new nozzle design; however, trip jet injection was discarded in favor of direct combustion with D_2 and F_2 until nozzle studies can be performed.

5.0 CONCLUSIONS

The direct injection of NF_3 into a stream of H atoms is insufficient at the temperature generated by reaction in the cavity. Combustion of NF_3 and D_2 provided an excellent source of NF_2 through control of the combustion mixture and thus the temperature. The production level of $N_2(B)$ implies that NF_2, and perhaps NF, was formed in the combustion. The lower $NF(a^1\Delta)$ production with NF_3 rather than with N_2F_4 is an unexpected result. This implies that NF is being formed in another state or that some $N(^2D)$ is being formed directly in the combustion. The large NH or ND peak seen when NF_3 is used may also indicate early $N(^2D)$ formation. Further exploration of the mechanism is required in order to explain these observations.

The question of whether complete mixing is occurrng is present in this set of experiments as was reported with the N_2F_4 studies (Ref. 9). Detailed hot flow mixing studies will be required to answer the level of mixing question.

These experiments are only intended to be preliminary studies to determine the utility of using NF_3 in place of N_2F_4 in $N_2(A)$ production. The results here indicate that the approach is promising. Further work must be accomplished in the areas of kinetics, mechanism and reactive flow mixing; however, it appears as if with some improvements $N_2(A)$ production levels needed for energy transfer could be achieved.

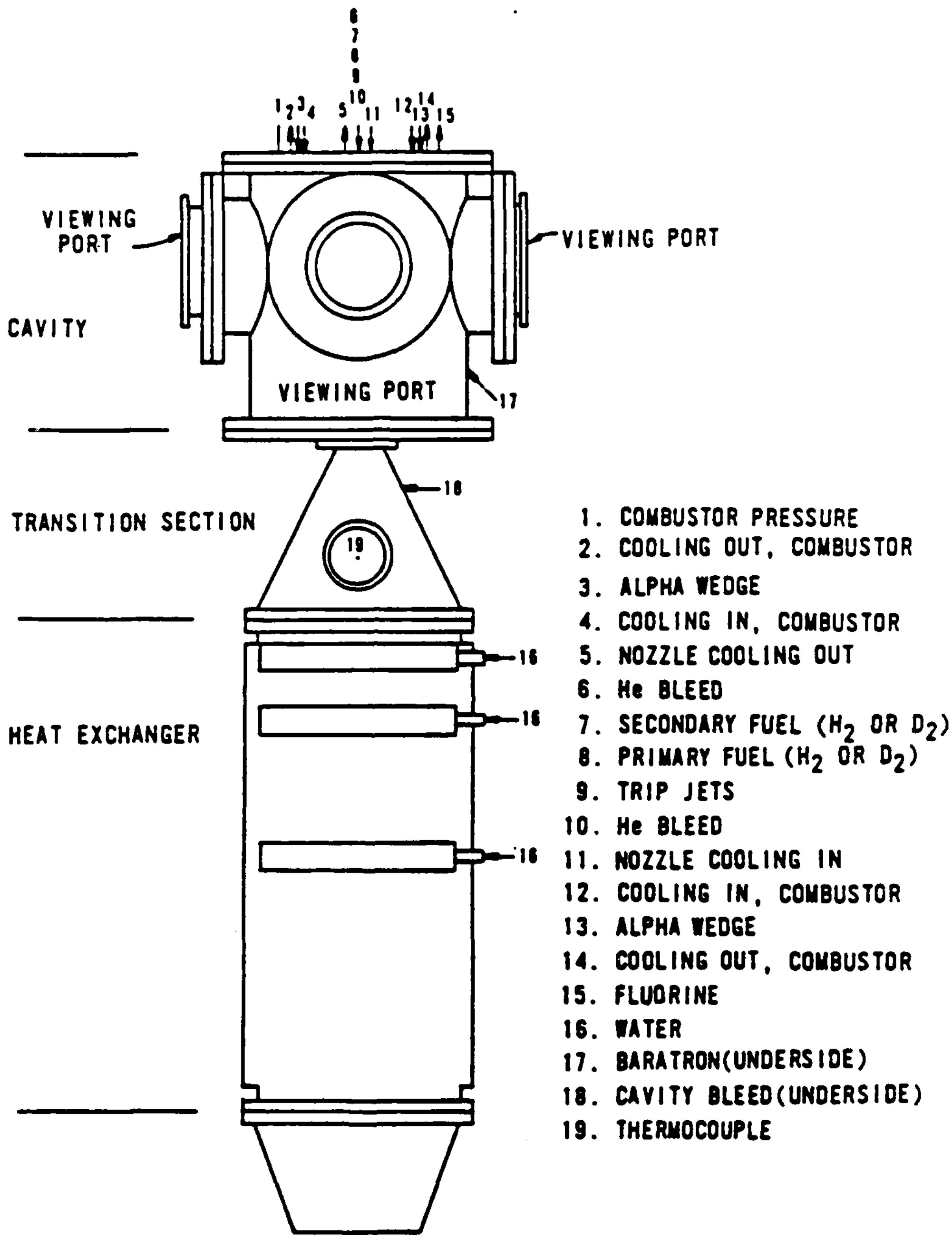


Figure 1. Device Schematic and Flow Input.

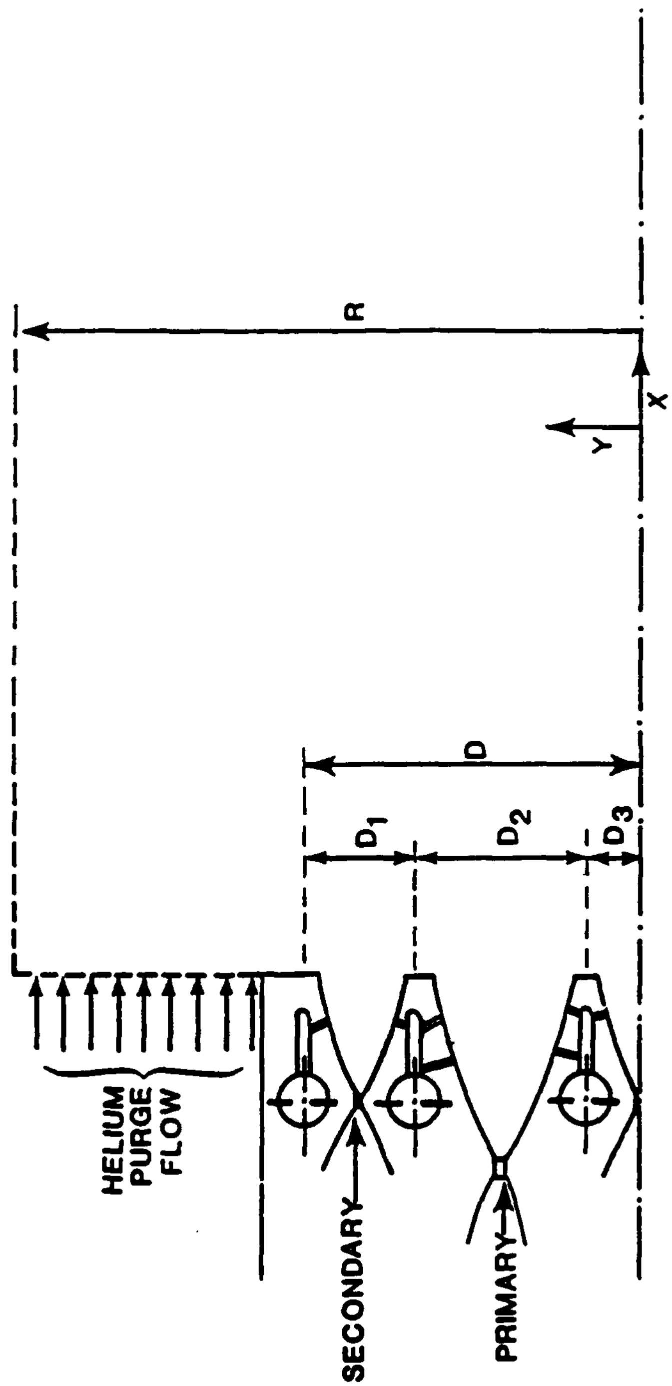


Figure 2. Half cross section of the BCL-16 nozzle.

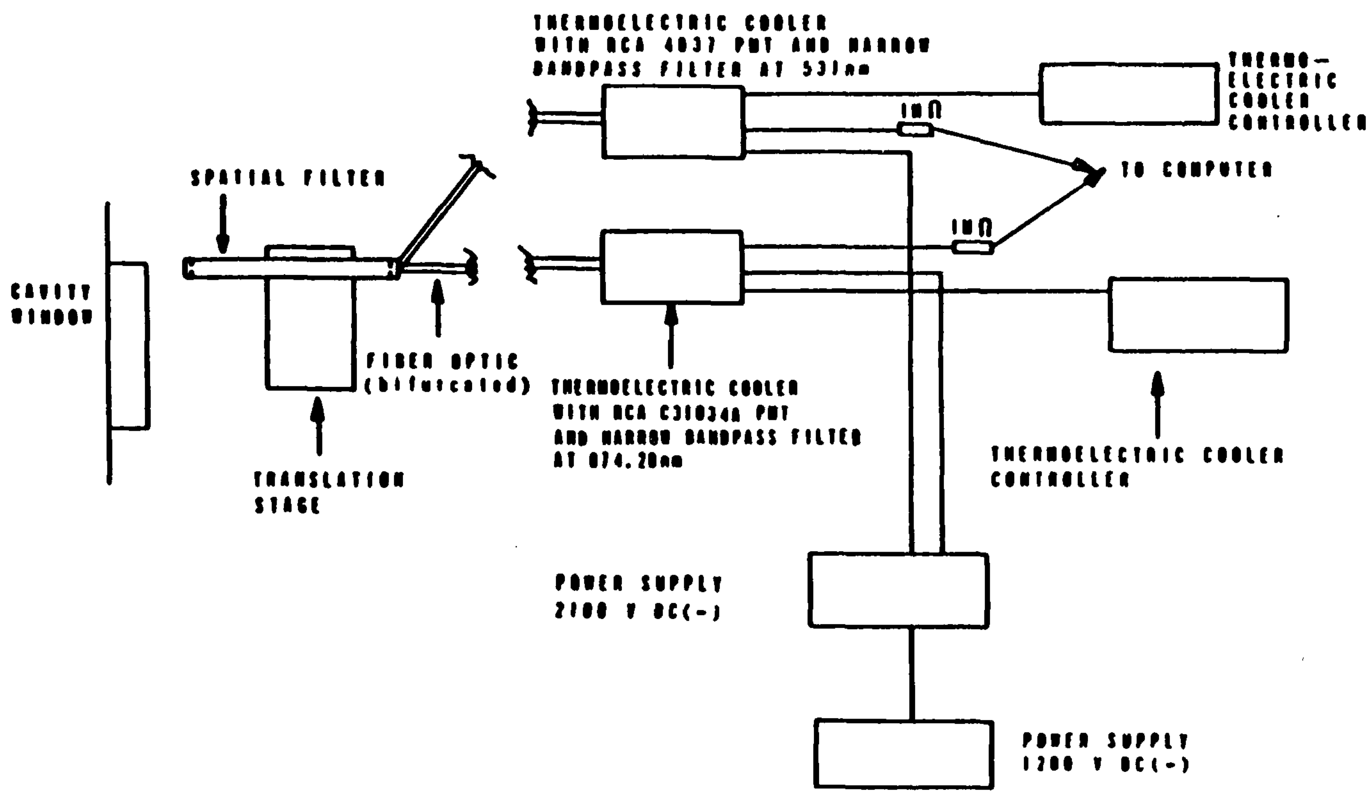


Figure 3. Schematic of the NF($a^1\Delta$) and NF($b^1\Sigma$) diagnostics.

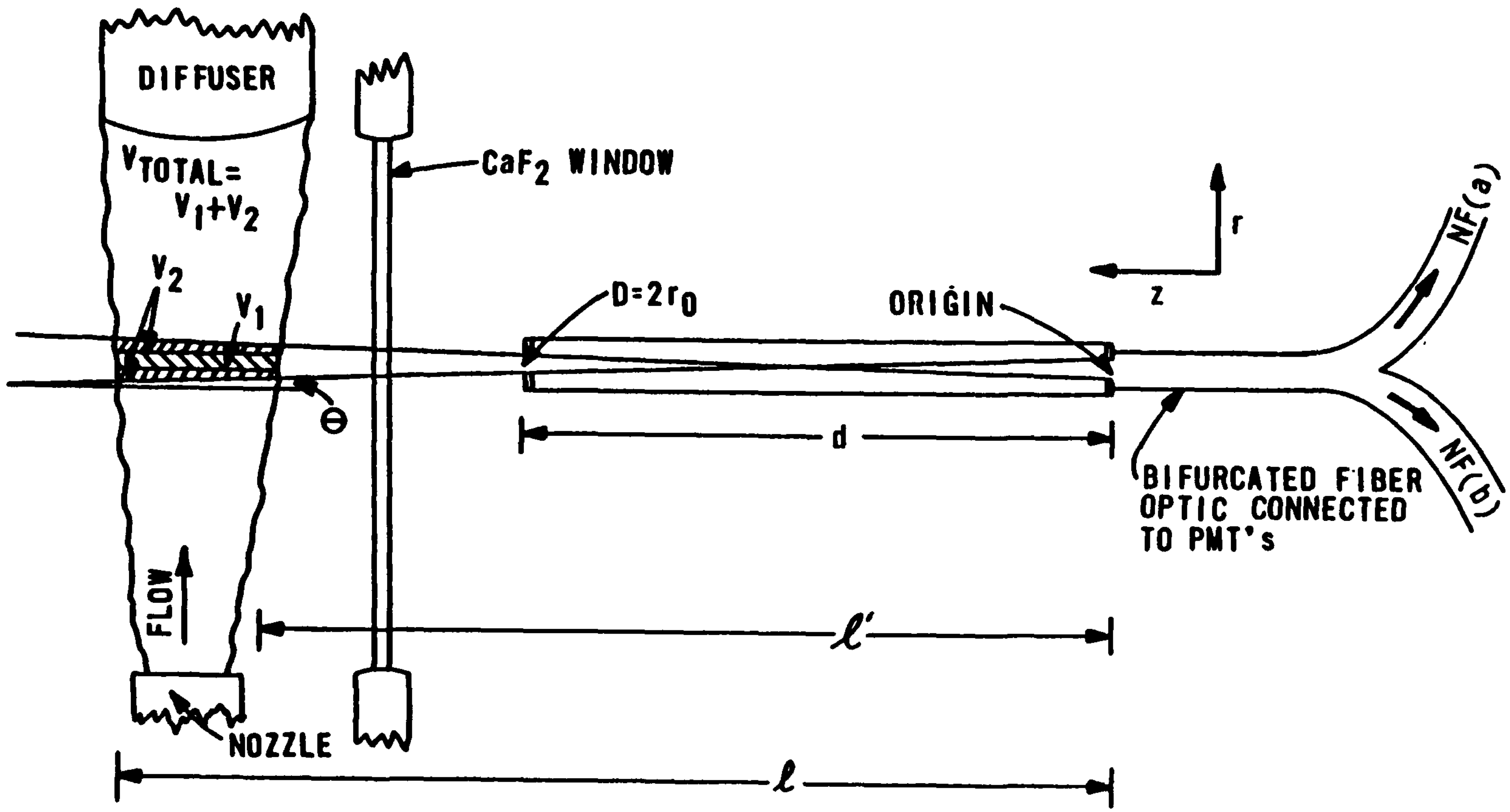


Figure 4. Experimental arrangement of the diagnostics.

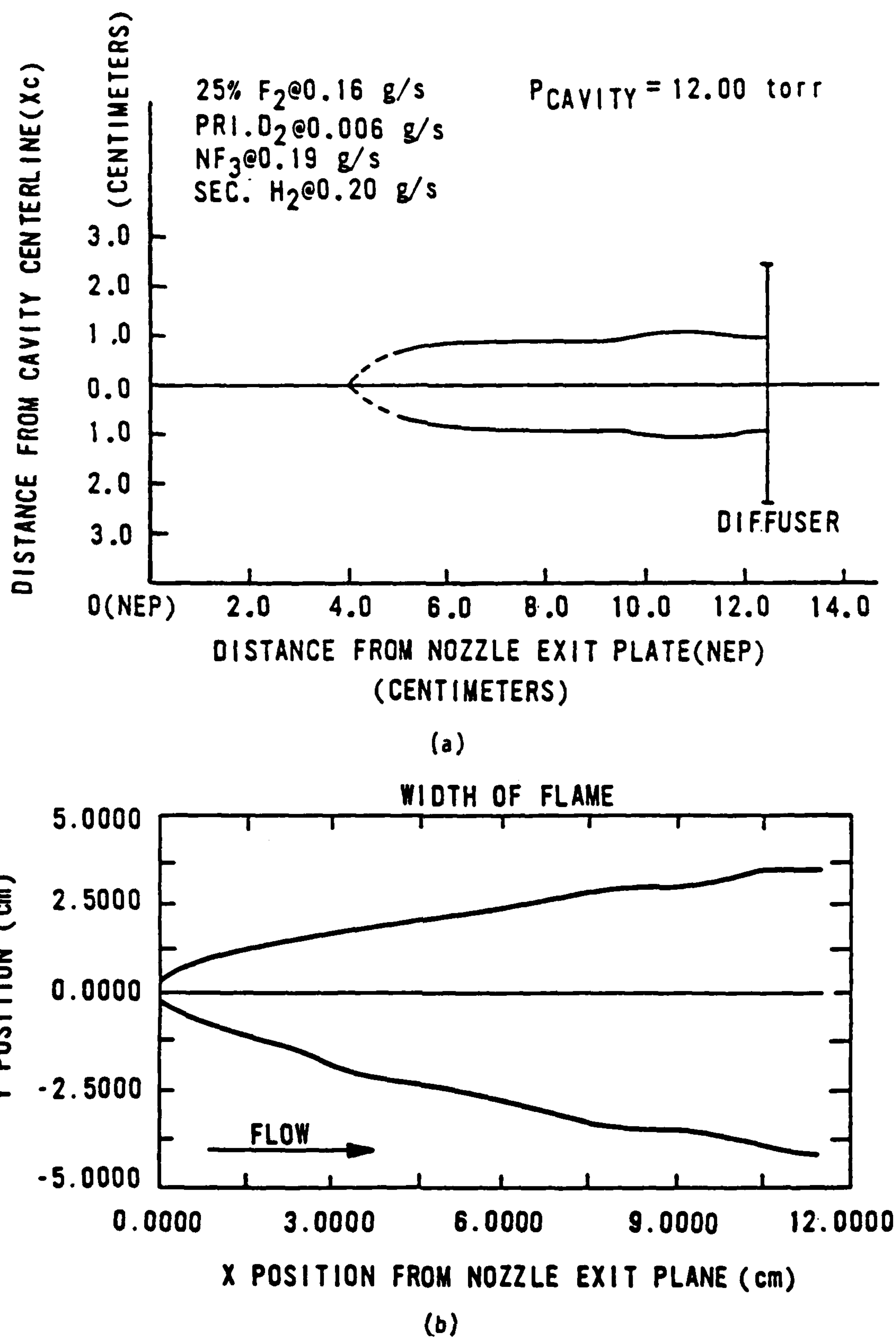


Figure 5. Digitized photograph of the flame (a) with NF_3 and (b) with N_2F_4.

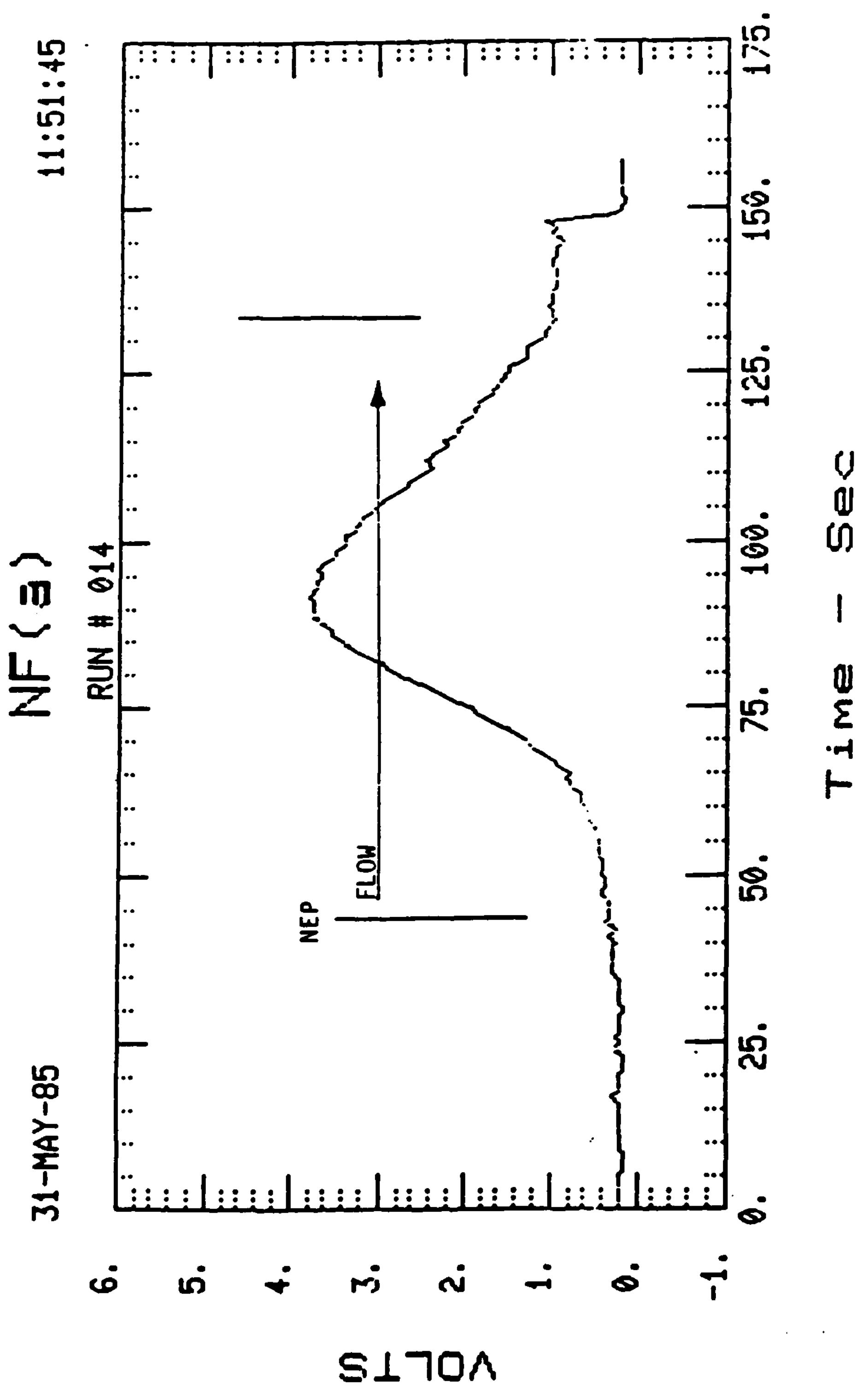


Figure 6. $NF(a^1\Delta)$ sample scan.

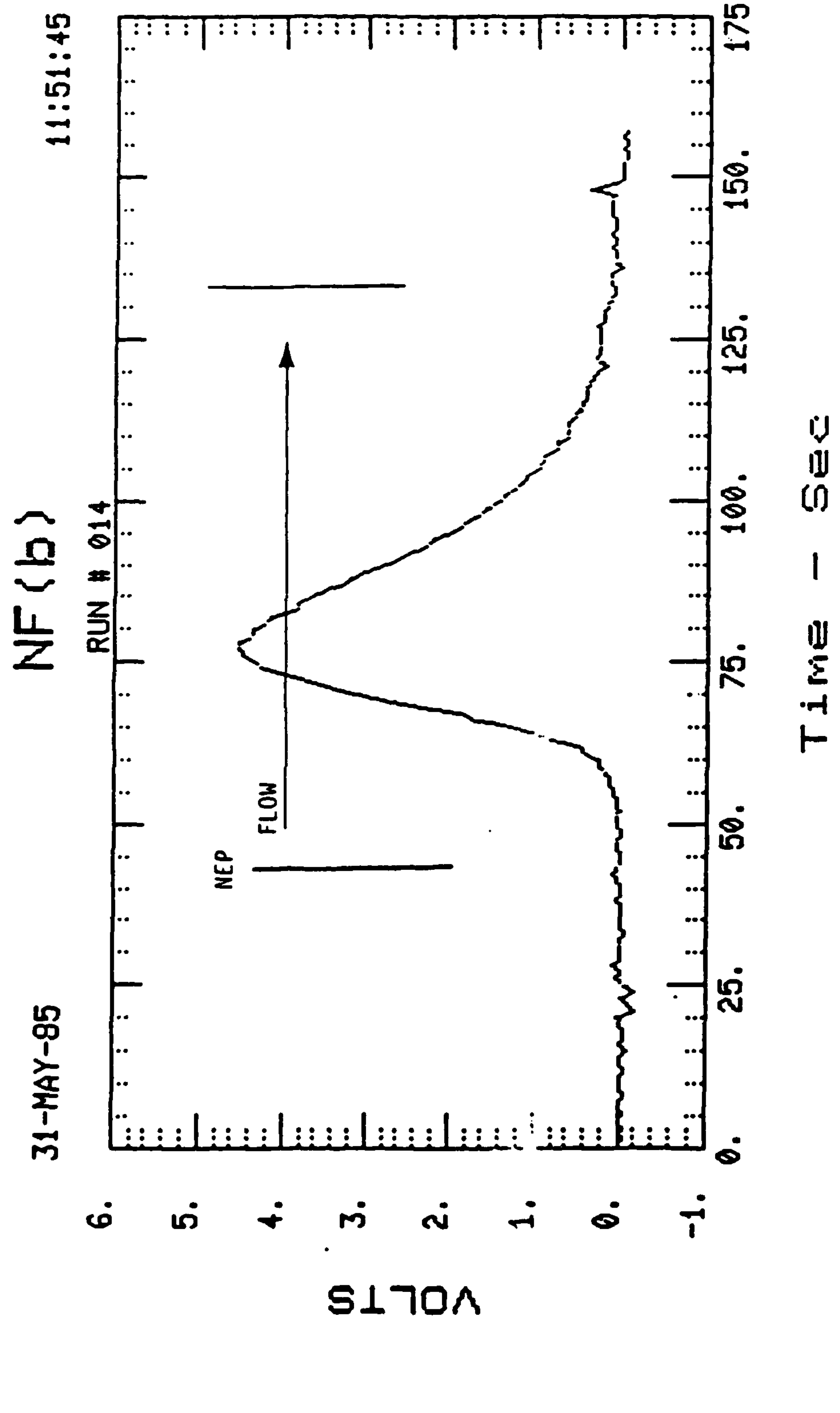
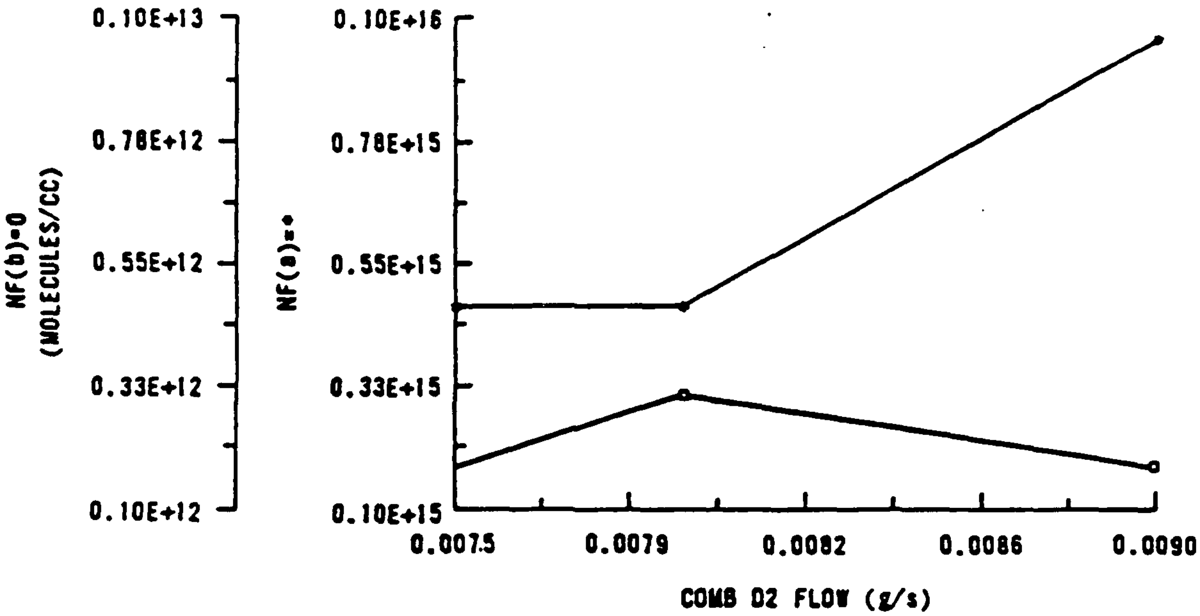


Figure 7. $NF(b^1\Sigma)$ sample scan.



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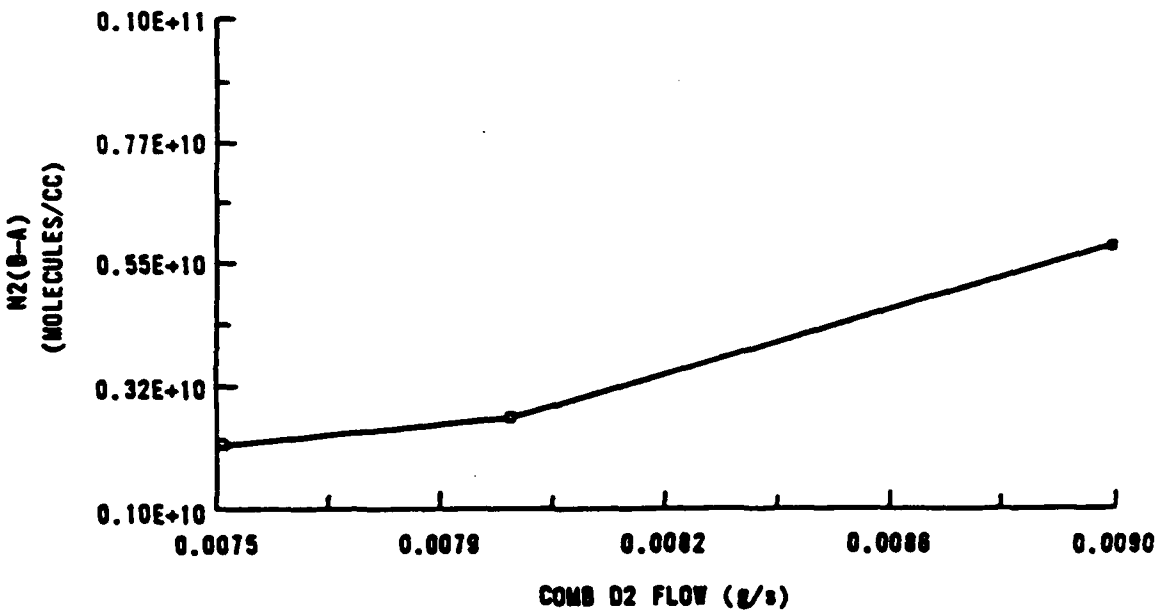
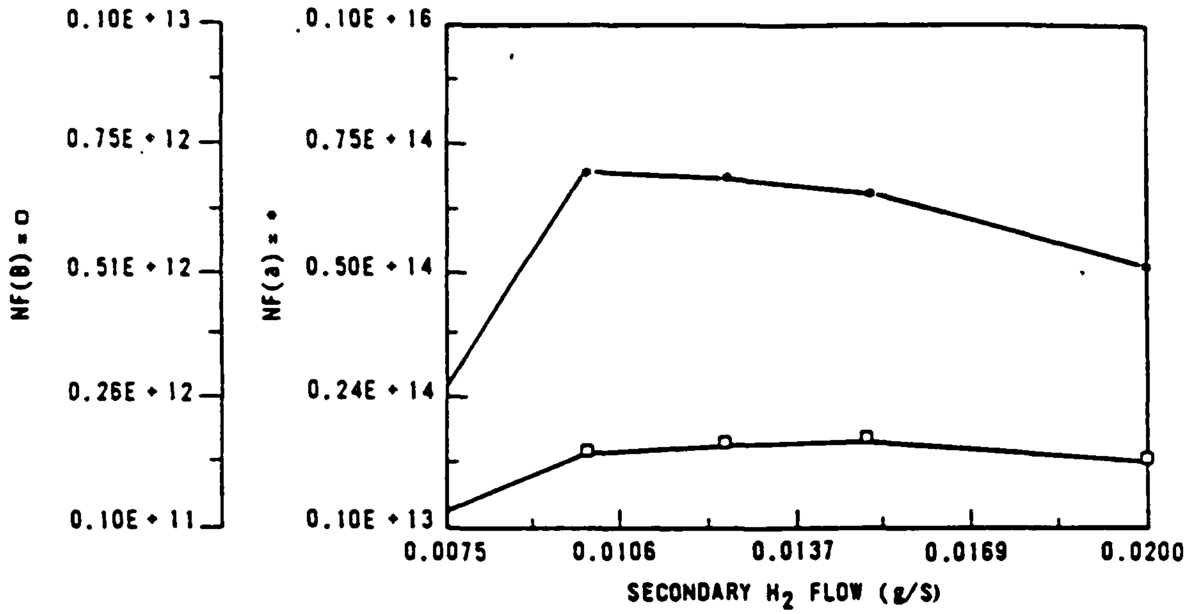


(b)

Figure 8. Species variation with D_2 combustor flow.



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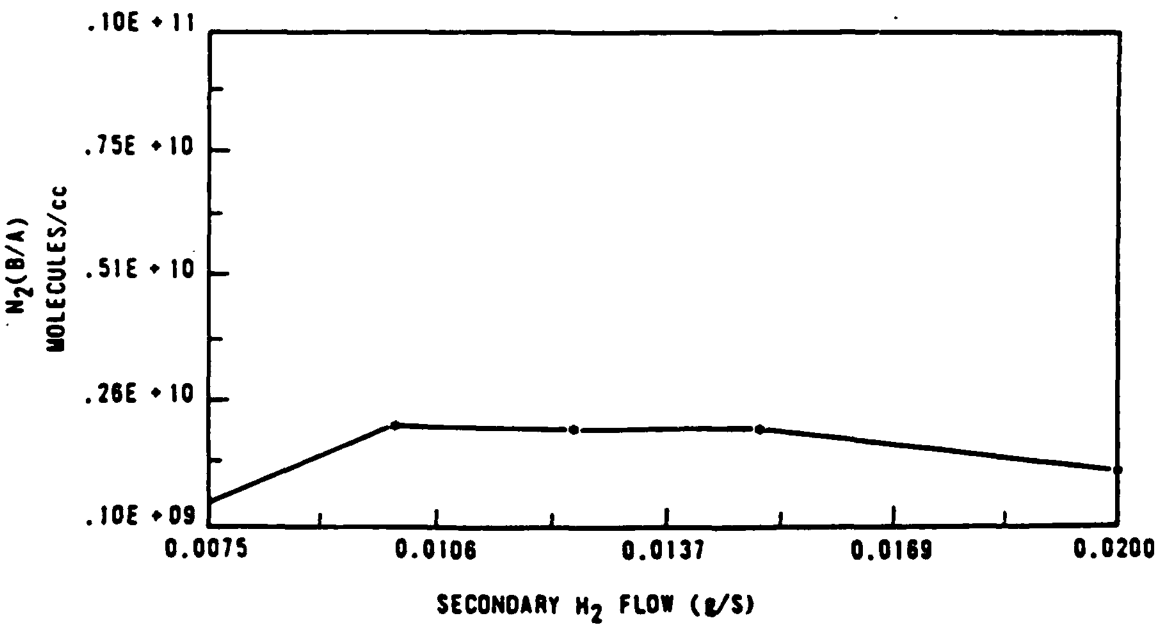

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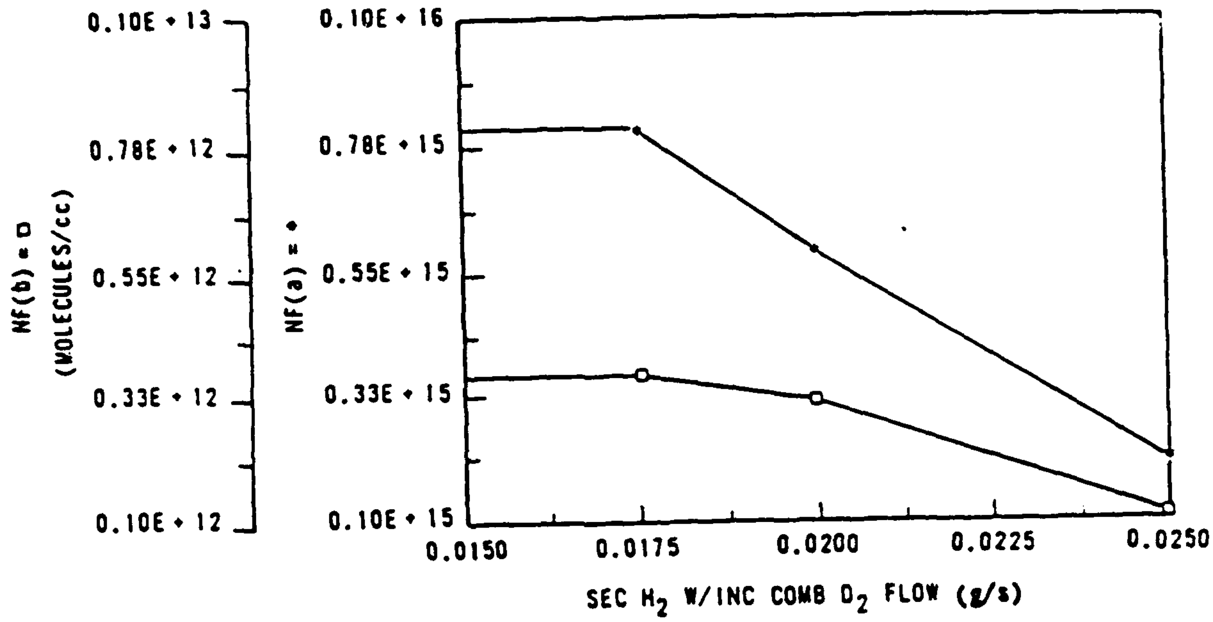
Figure 9. Species variation with H_2 secondary flow.



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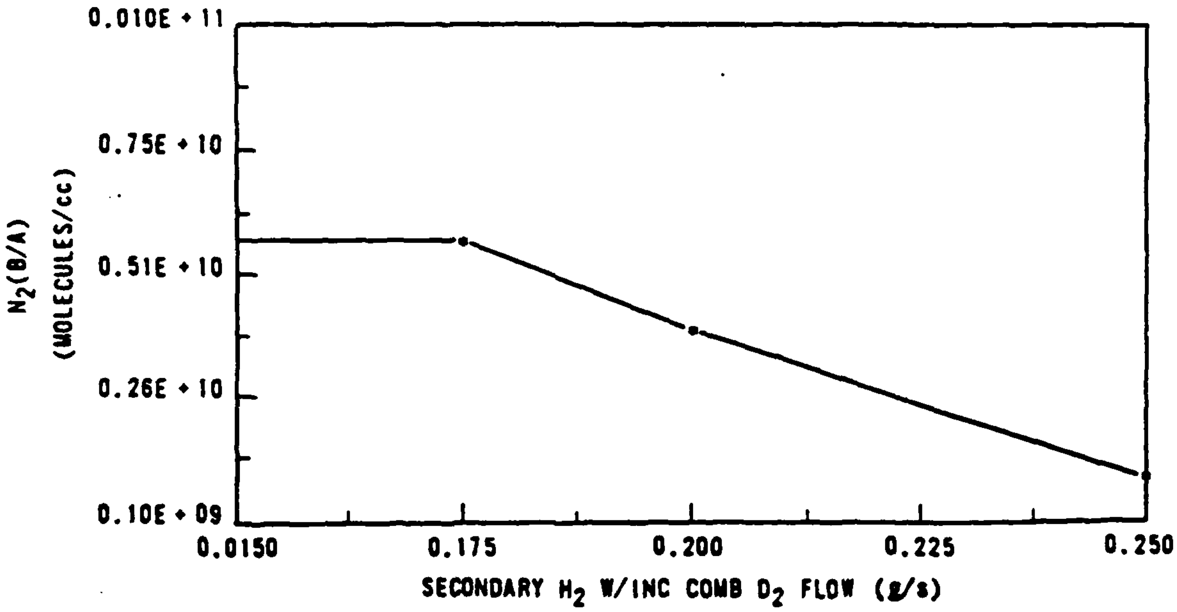
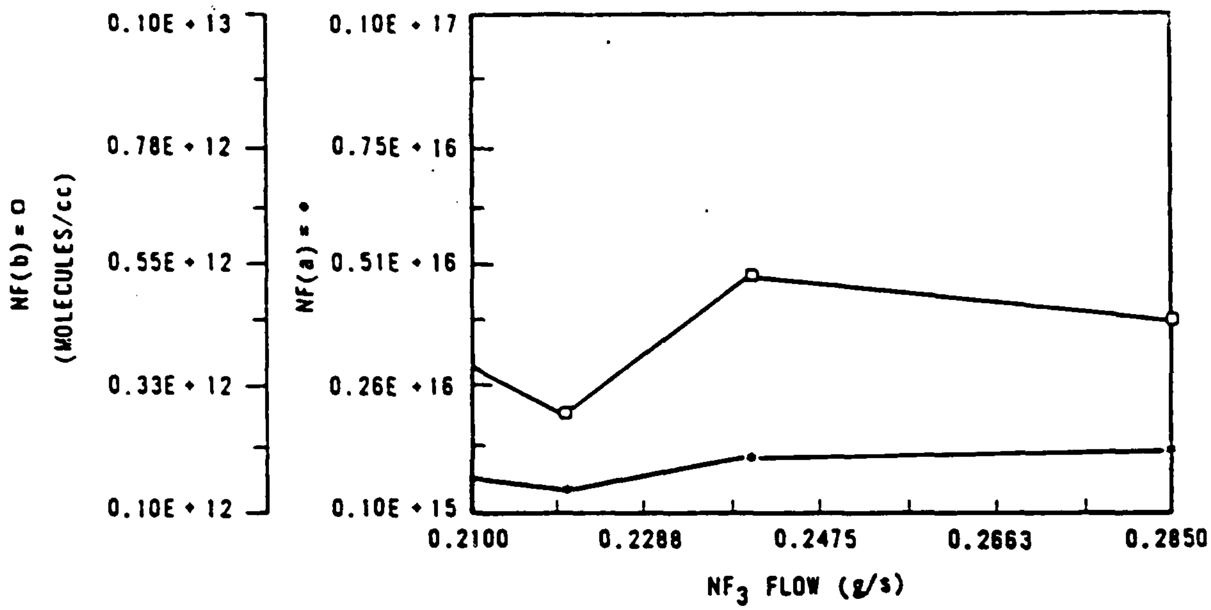


(b)

Figure 10. Species variation, at increased combustor D_2, with H_2 secondary flow.



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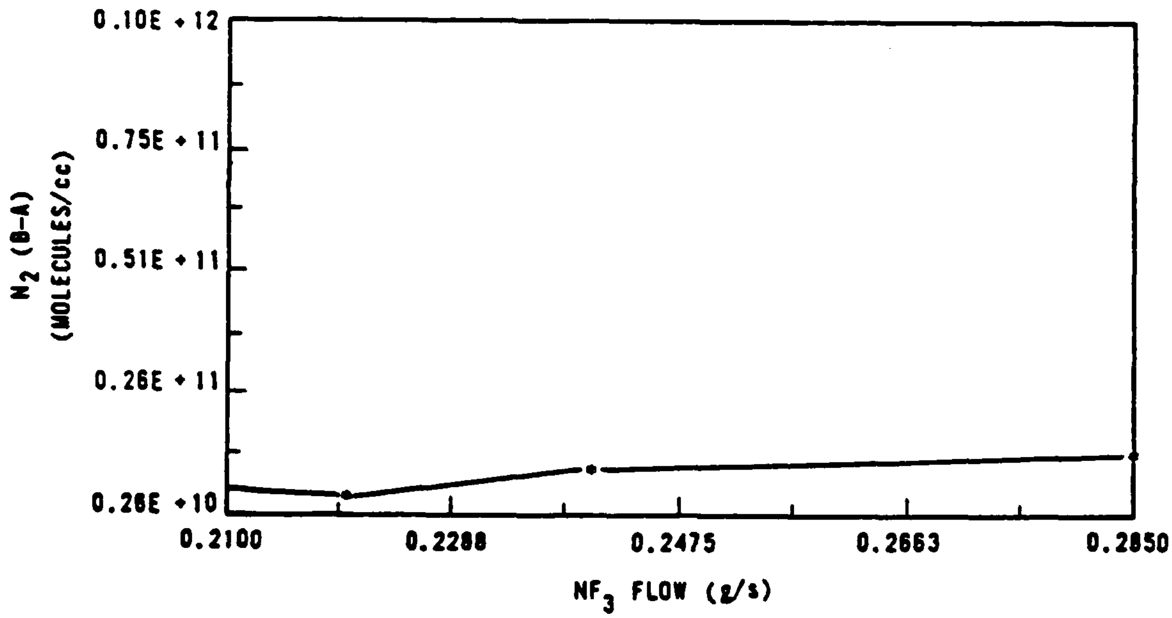

(b)

Figure 11. Species variation with NF_3 flow.

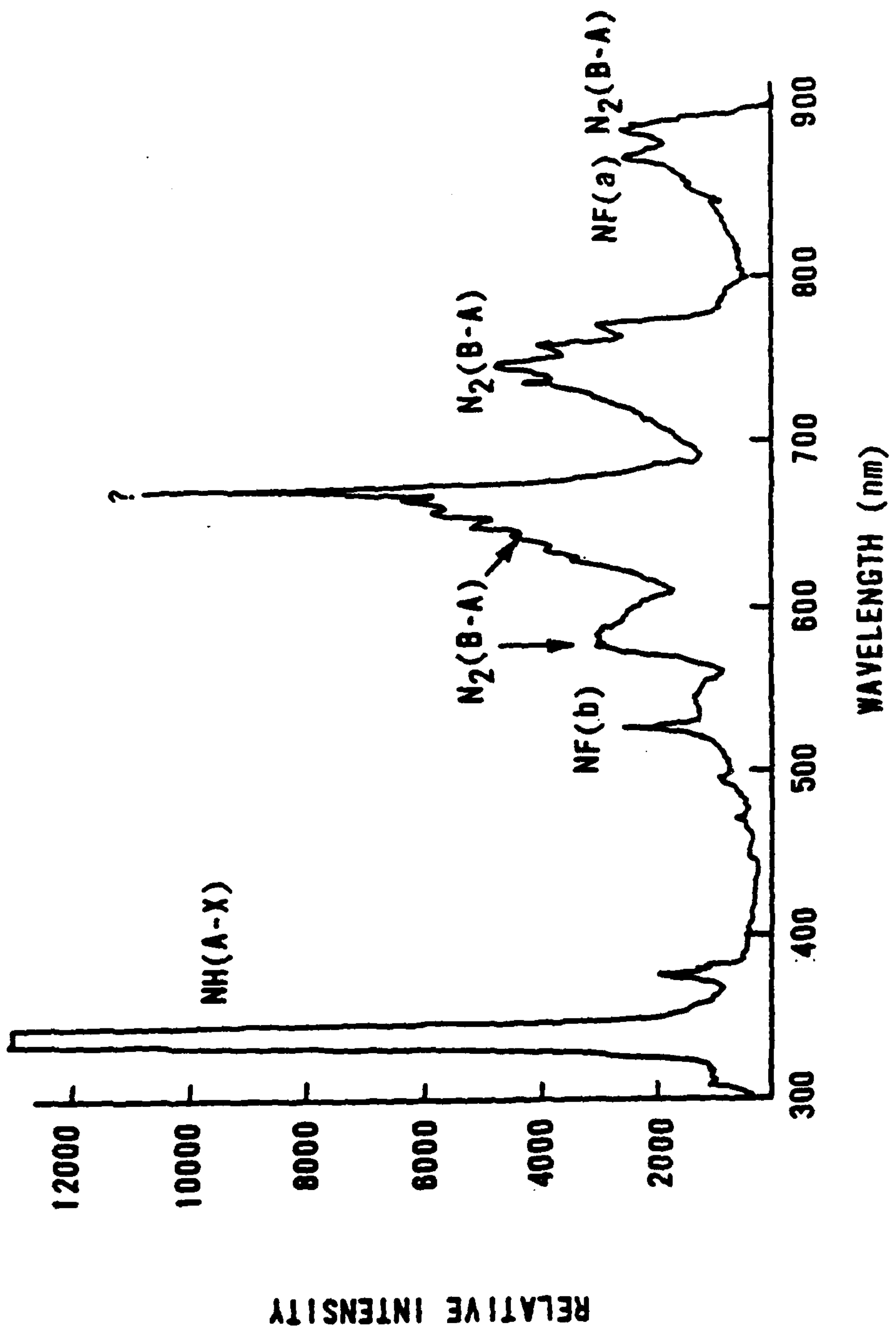

Figure 12. OMA III scan (uncorrected) of flow with NF_3.

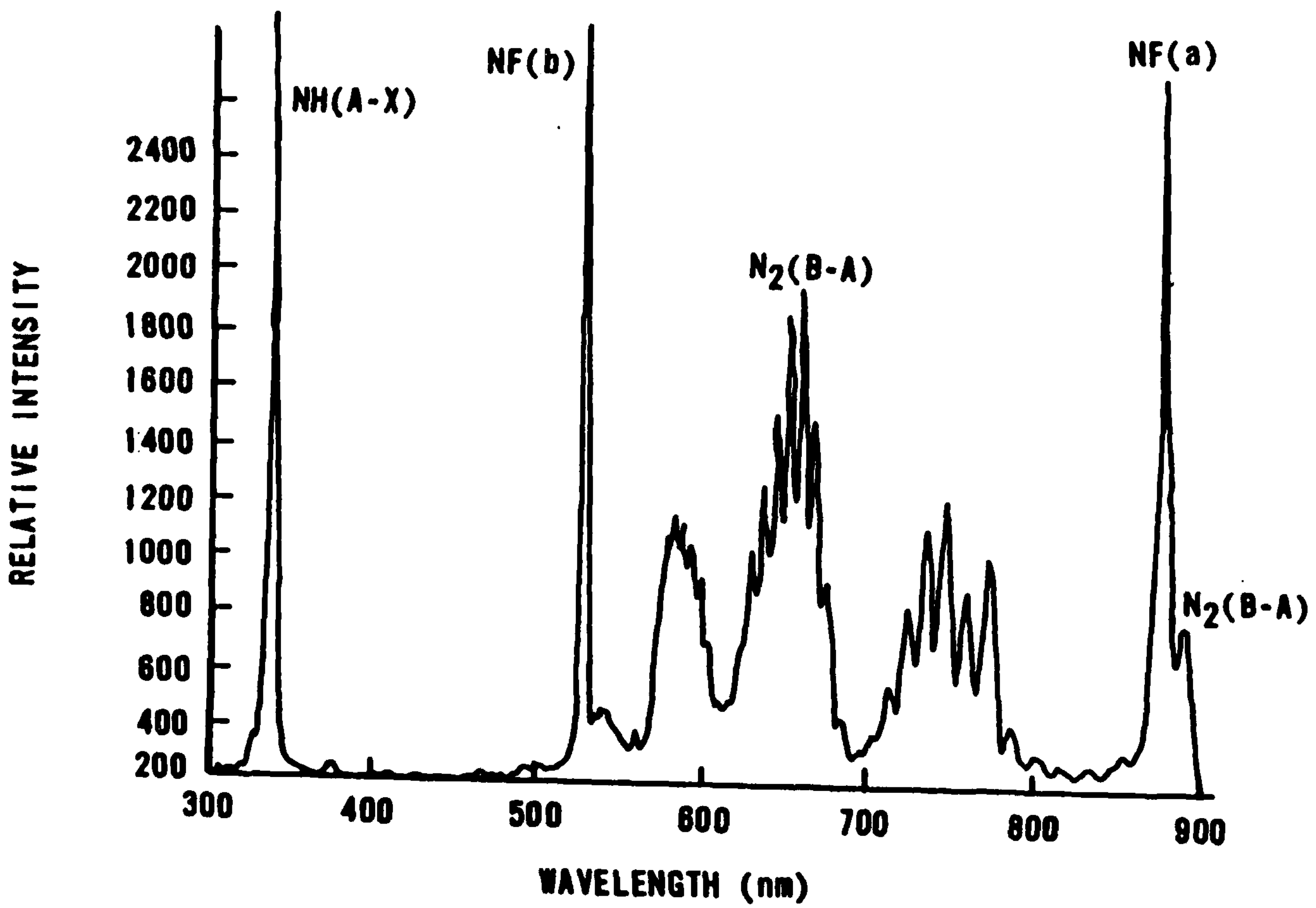

Figure 13. OMA III scan (uncorrected) of flow with N_2F_4.

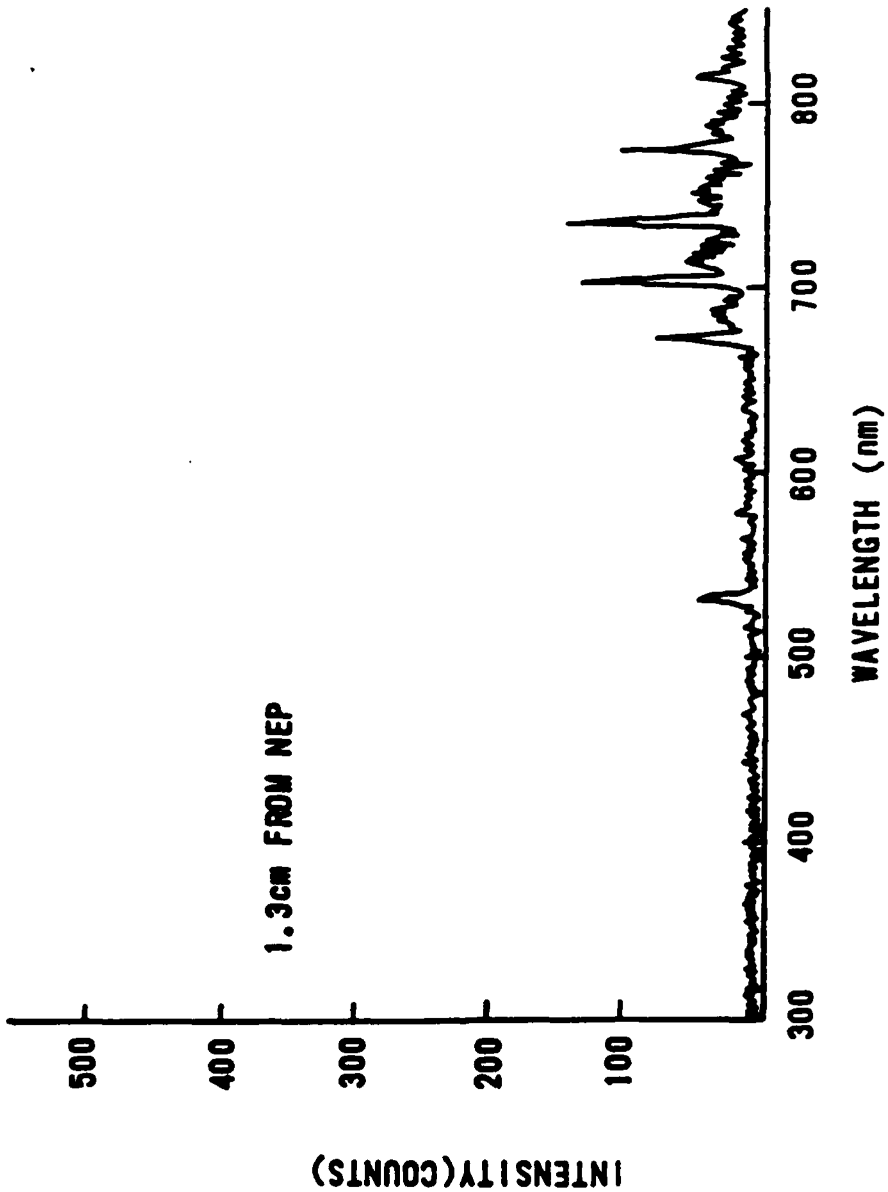

Figure 14. OMA III scan at 1.3 cm.

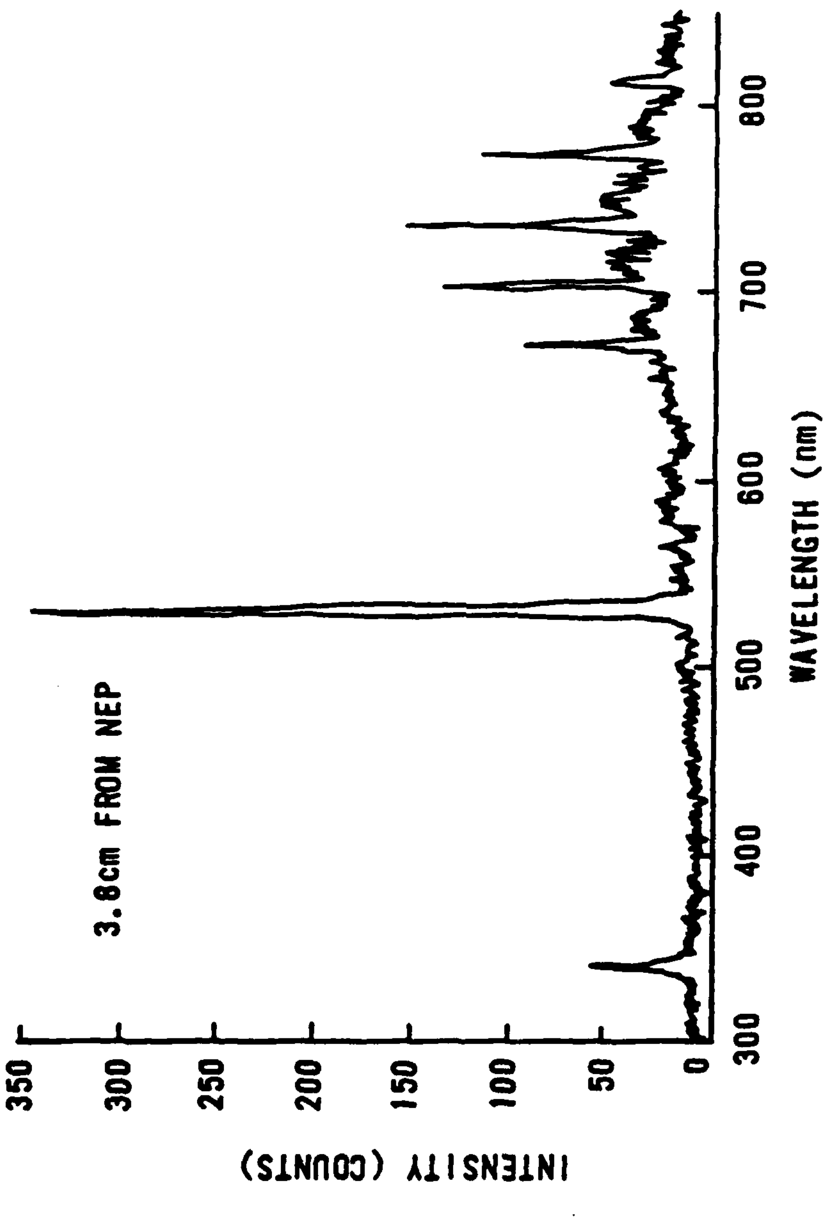

Figure 15. OMA III scan at 3.8 cm

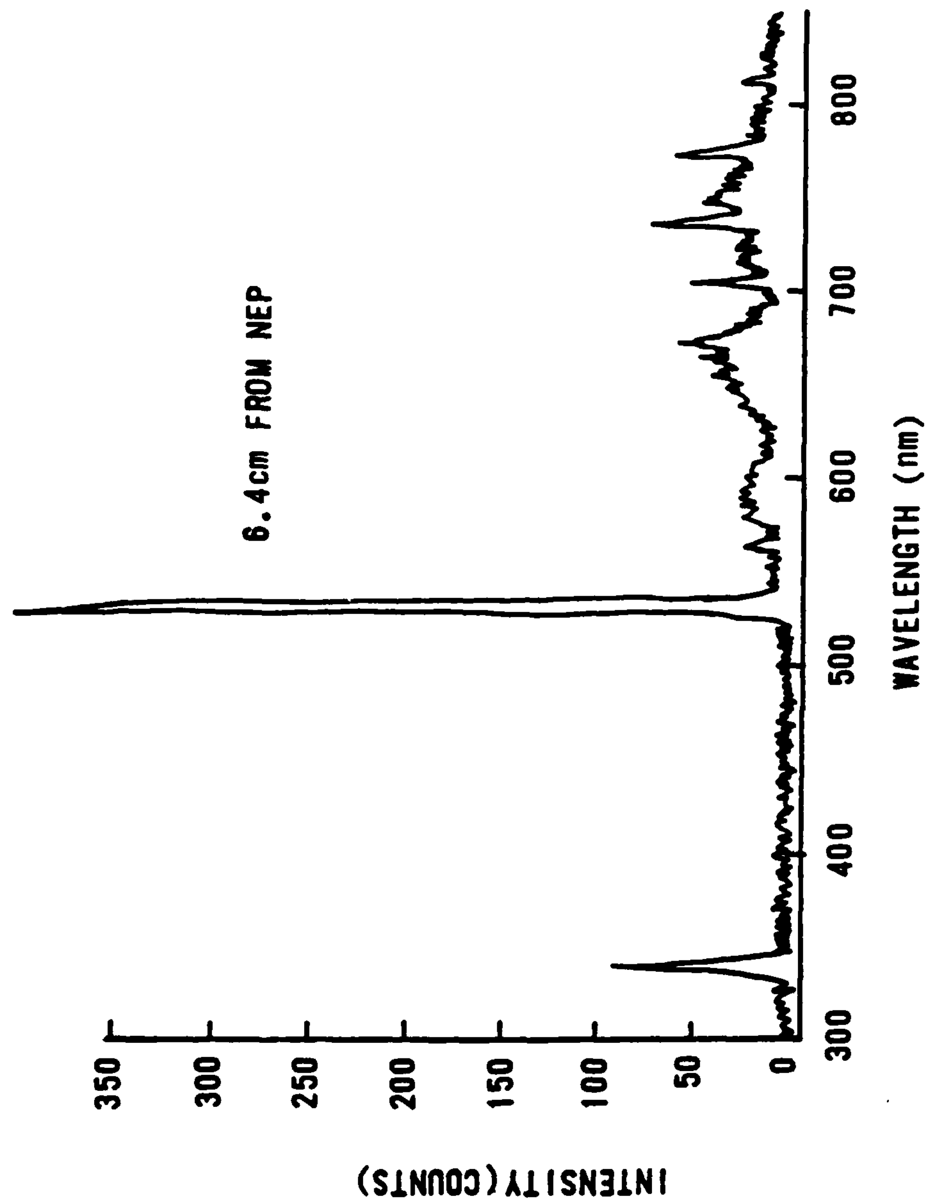

Figure 16. OMA III scan at 6.4 cm.

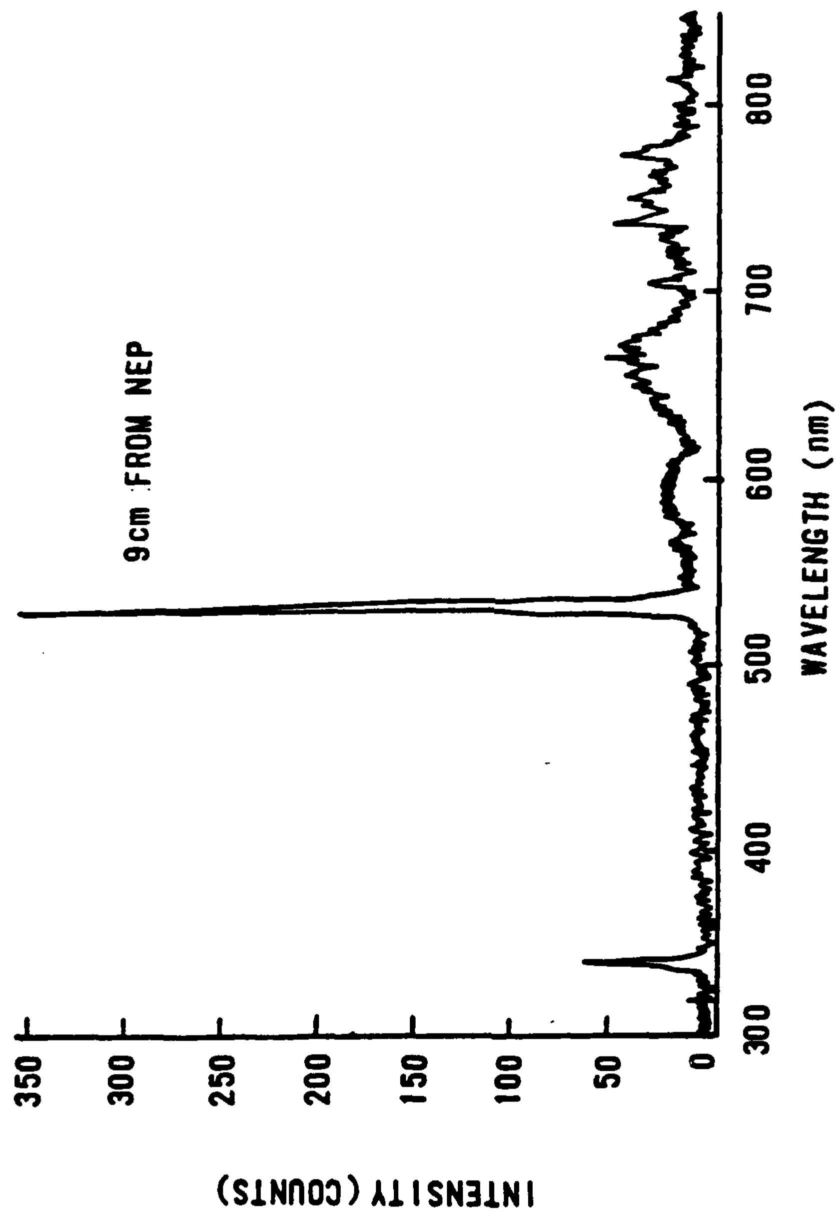

Figure 17. OMA III scan at 9 cm.

TABLE 1. NF_3 COMBUSTION DATA

Test-Run	Primary (g/s)		Secondary (g/s)			P_{cav} (torr)[a]	(molecules/cm^3)		
	25% F_2 in He	NF_3	D_2	H_2	He		$NF(a^1\Delta)$	$NF(b^1\ )$	$N_2(B)$
19-9	0.15	0.14	0.006	0.011	0	12.0	3.1×10^{15}	1.0×10^{12}	2.2×10^{10}
19-12	0.15	0.21	0.006	0.015	0	12.0	4.5×10^{15}	1.6×10^{12}	3.6×10^{10}
19-15	0.15	0.24	0.006	0.023	0	12.0	7.1×10^{15}	2.2×10^{12}	5.4×10^{10}
20-10	0.14	0.20	0.006	0.015	0	12.0	6.6×10^{14}	1.8×10^{11}	1.39×10^{9}
20-13	0.14	0.23	0.009	0.015	0	12.0	1.2×10^{15}	5.3×10^{11}	6.5×10^{9}
20-15	0.16	0.27	0.010	0.015	0	12.0	1.7×10^{15}	6.2×10^{11}	8.5×10^{9}

[a] torr = 1.33×10^2 pascal

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